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An analysis of dynamic roughening and smoothening mechanisms of thin films grown with pulsed-dc magnetron sputtering is presented. The roughness evolution has been described by a linear stochastic equation, which contains the second- and fourth-order gradient terms. Dynamic smoothening of the growing interface is explained by ballistic effects resulting from impingements of ions to the growing thin film. These ballistic effects are sensitive to the flux and energy of impinging ions. The predictions of the model are compared with experimental data, and it is concluded that the thin film roughness can be further controlled by adjusting waveform, frequency, and width of dc pulses. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3037237]

I. INTRODUCTION

Nanocomposite thin films based on amorphous diamondlike carbon (DLC) have become increasingly important for advanced applications because of the unique combination of structural and physical properties. Amorphous DLC thin films with various composition and physical properties can be produced by different deposition techniques, such as plasma assisted chemical-vapor deposition, magnetron sputtering, ion beam deposition, laser ablation, and vacuum cathodic arc depositions.[1] The common feature of the most of these techniques is the intense impingement of the growing film by energetic ions.[1–3] For many applications, atomically flat surfaces of thin films are desired. Studies of ion beam deposition of carbon ions have found that the roughness varies in a consistent way with the ion energy,[4–6] so that a minimum roughness is obtained at the ion energy of about 100 eV, which corresponds to the maximum sp^3 fraction (for references see the review in Ref. 1). According to the models proposed up to date, the formation of sp^3 bonds is controlled by subplantation of energetic carbon atoms just below the growing interface.[7–9]

Surface roughness of DLC films prepared using (i) rf glow discharge from methane, (ii) dc magnetron sputtering of a graphite target, with a rf substrate bias, and (iii) an ion beam generated from a cathodic arc discharge has been studied in Refs. 10 and 11. It has been found that below 50 eV ion energy of impingement the growing interface was very rough on a nanoscale for all the three techniques studied, while above that threshold value the roughness dropped sharply. This value is close to the threshold energies of atomic displacement E_d=37–47 eV in graphite and diamond.[1] These observations had been predicted by the molecular dynamics (MD) simulation of deposition processes involving subplantation,[8] which showed that surface processes are dominant for deposition at impinging energy below 30 eV, while subsurface processes govern the structure formation at higher energies of ion impingement.

The aim of this paper is to understand the mechanisms of dynamic smoothening of the growing interface and to reveal the parameters and processes that control the evolution of interface roughness in thick films. In Sec. II we present a phenomenological model of roughening and the experimental data are analyzed accordingly. For details of the closed-field unbalanced magnetron sputtering system, reference is made to our previous work.[12]

II. THE MODEL

A. Mathematical description of roughening kinetics

Several models of roughening kinetics and the scaling behavior of random surface structures during film growth are reviewed in Refs. 13–15. In particular, kinetic Monte Carlo models are frequently used for the simulation of film growth on an atom-by-atom basis using probabilistic rules to govern deposition, diffusion, and other growth processes. The drawback of this approach is that it requires a detailed knowledge about all processes taking place on an atomic scale. In this paper we propose a continuum approach that is based on a stochastic differential equation. The continuum models proved to be useful for the understanding of roughening kinetics, despite the complexity of the growth process on atomic scale.

Since the roughness of the thin films deposited in our experiments is very low[12] we will consider a "minimal" equation of interface motion (without nonlinear terms), which contains the Edwards–Wilkinson term $D_2\nabla^2(\ldots)$ (Ref. 16) and the Mullins term $D_4\nabla^2[\nabla^2(\ldots)]$,[17]

$$\frac{\partial h(\mathbf{r},t)}{\partial t} = D_2\nabla^2 h(\mathbf{r},t) - D_4\nabla^2[\nabla^2 h(\mathbf{r},t)] + \eta(\mathbf{r},t), \qquad (1)$$

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$$h(\mathbf{r},0) = h_0(\mathbf{r}), \tag{2}$$

where D_2 and D_4 are the positive diffusivity parameters that control the atomic mobility along the interface. $h(\mathbf{r},t)$ is the deviation of the interface profile $Z(\mathbf{r},t)=h(\mathbf{r},t)+Ft$ from the flat interface of the growing film of mean thickness $\langle Z(\mathbf{r},t)\rangle = Ft$ and $h_0(\mathbf{r})$ is the initial (substrate) profile. The stochastic noise term $\eta(\mathbf{r},t)$ is related to the fluctuations of deposition flux and is assumed to be Gaussian and uncorrelated with zero mean. The noise covariance is given by

$$\langle \eta(\mathbf{r},t)\eta(\mathbf{r}',t')\rangle = D\delta(\mathbf{r}-\mathbf{r}')\delta(t-t'), \tag{3}$$

where $\langle \ldots \rangle$ stands for the ensemble average and $\delta(\mathbf{r}-\mathbf{r}')$ is the two-dimensional delta function.

The scaling behavior of Eq. (1) in various limiting cases has been studied in Ref. 18. The diffusion terms in Eq. (1) become comparable at the length scale

$$L^* = \sqrt{\frac{D_4}{D_2}}. \tag{4}$$

Therefore the smoothening process is governed by the fourth derivative term on scales smaller than L^*, the second order Edwards–Wilkinson term dominates on scales larger than L^*.

Here we will focus on the smoothening of initially rough surface. Our goal is to solve Eq. (1) to fit the solution to the experimental data and to evaluate the parameters D, D_2, and D_4. Applying the Fourier transform

$$f(\mathbf{q}) = \int h(\mathbf{r})\exp(-i\mathbf{q}\mathbf{r})d\mathbf{r}, \tag{5}$$

we find that Fourier components of the height evolve independently according to

$$\frac{\partial f(\mathbf{q},t)}{\partial t} = -a(q)f(\mathbf{q},t) + \phi(\mathbf{q},t), \tag{6}$$

where

$$a(q) = D_2q^2 + D_4q^4, \tag{7}$$

$$\phi(\mathbf{q},t) = \int \eta(\mathbf{r},t)\exp(-i\mathbf{q}\mathbf{r})d\mathbf{r}. \tag{8}$$

The Fourier transform of the noise has the following correlation properties:

$$\langle \phi(\mathbf{q},t)\phi^*(\mathbf{q}',t')\rangle = 4\pi^2 D\delta(\mathbf{q}-\mathbf{q}')\delta(t-t'). \tag{9}$$

Solution of Eq. (6) consists of contributions due to (i) initial profile $f_i(\mathbf{q},t)$ and (ii) the deposition noise $f_\eta(\mathbf{q},t)$,

$$f(\mathbf{q},t) = f_i(\mathbf{q},t) + f_\eta(\mathbf{q},t) = f_0(\mathbf{q})e^{-a(q)t} + \int_0^t \phi(\mathbf{q},t')e^{-a(q)(t-t')}dt', \tag{10}$$

where $f_0(\mathbf{q})$ is the Fourier transform of the initial profile.

The interface evolution at a moment t can be described by the correlation function

$$C(\mathbf{r},t) = \langle h(\mathbf{r}',t)h(\mathbf{r}'+\mathbf{r},t)\rangle, \tag{11}$$

where $h(\mathbf{r},t)$ is the sum of two terms $h(\mathbf{r},t)=h_i(\mathbf{r},t)+h_\eta(\mathbf{r},t)$ according to Eq. (10). Since the initial roughness is not correlated with the deposition process, the cross-correlation term in Eq. (11) is zero, namely, $\langle h_i(\mathbf{r}',t)h_\eta(\mathbf{r}'+\mathbf{r},t)\rangle = 0$. Therefore, the correlation function can be represented as a sum of two terms averaged over the initial roughness $\langle \ldots \rangle_{h_0}$ and the deposition noise $\langle \ldots \rangle_\eta$, respectively,

$$C(\mathbf{r},t) = C_i(\mathbf{r},t) + C_\eta(\mathbf{r},t) = \langle h_i(\mathbf{r}',t)h_i(\mathbf{r}'+\mathbf{r},t)\rangle_{h_0} + \langle h_\eta(\mathbf{r}',t)h_\eta(\mathbf{r}'+\mathbf{r},t)\rangle_\eta. \tag{12}$$

Using Eq. (10), we find the correlation function for the initially flat surface

$$C_\eta(\mathbf{r},t) = \langle h_\eta(\mathbf{r}',t)h_\eta(\mathbf{r}'+\mathbf{r},t)\rangle_\eta = \frac{1}{(2\pi)^4}\int d\mathbf{q}\int d\mathbf{q}'\langle f_\eta(\mathbf{q}',t)f_\eta^*(\mathbf{q},t)\rangle \times e^{-i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{r}'+i\mathbf{q}'\cdot\mathbf{r}}, \tag{13}$$

where

$$\langle f_\eta(\mathbf{q}',t)f_\eta^*(\mathbf{q},t)\rangle = (2\pi)^2 S_\eta(\mathbf{q},t)\delta(\mathbf{q}-\mathbf{q}') \tag{14}$$

and $S_\eta(\mathbf{q},t)$ is the power spectrum distribution (PSD) of the film grown on the initially flat substrate

$$S_\eta(\mathbf{q},t) = \frac{D}{2a(q)}(1-e^{-2a(q)t}). \tag{15}$$

Consider the contribution to the correlation function due to the initial profile

$$C_i(\mathbf{r},t) = \frac{1}{(2\pi)^4}\exp\{-[a(q)+a(q')]t\} \times \int\int \langle f_0(\mathbf{q}')f_0^*(\mathbf{q})\rangle e^{-i\mathbf{q}\cdot\mathbf{r}'+i\mathbf{q}'\cdot(\mathbf{r}'+\mathbf{r})}d\mathbf{q}d\mathbf{q}'. \tag{16}$$

It is reasonable to assume that in the initial profile the different q modes are uncorrelated, i.e.,

$$\langle f_0(\mathbf{q}')f_0^*(\mathbf{q})\rangle = (2\pi)^2 S_0(\mathbf{q})\delta(\mathbf{q}-\mathbf{q}'), \tag{17}$$

where $S_0(\mathbf{q})$ is the PSD of the substrate.

Thus the final expression for the correlation function is given by

$$C(\mathbf{r},t) = \frac{1}{4\pi^2}\int S(\mathbf{q},t)e^{i\mathbf{q}\cdot\mathbf{r}}d\mathbf{q}, \tag{18}$$

$$S(\mathbf{q},t) = S_0(\mathbf{q})e^{-2a(q)t} + S_\eta(\mathbf{q},t). \tag{19}$$

It is seen that the contribution to the correlation function due to the initial profile is a decreasing function of time. If the statistical properties of the rough interface are isotropic then $C(\mathbf{r})$ is a function of $r=|\mathbf{r}|$.

$$C(r,t) = \frac{1}{2\pi}\int S(q,t)J_0(qr)qdq, \tag{20}$$

where $J_0(qr)$ is the Bessel functions of the first kind.

Experimentally, the interface profile is measured in discrete points $N\times N$ for the sample size $L\times L$; therefore, to compare with the experimental data we will use the discrete Fourier transform for the set of height data points $h_{kl}=h(x_k,y_l)$.

$$g_{nm}=\frac{1}{N^2}\sum_{k,l=0}^{N-1}h_{kl}\exp\left(-2\pi i\frac{kn+lm}{N}\right). \tag{21}$$

The continuous Fourier transform and the discrete one are related by

$$f(\mathbf{q})=f(q_x,q_y)\Leftrightarrow L^2 g_{nm}, \tag{22}$$

where $q_x=2\pi n/L$ and $q_y=2\pi m/L$; $n, m<N$.

For samples sufficiently large to represent statistical properties of the thin film interface, the ensemble average over initial roughness and deposition noise is equivalent to the averaging over spatial coordinates

$$C(\mathbf{r},t)=\frac{1}{L^2}\int_{-L/2}^{L/2}\int_{-L/2}^{L/2}h(\mathbf{r}',t)h(\mathbf{r}'+\mathbf{r},t)d\mathbf{r}'$$

$$\approx\frac{1}{4\pi^2L^2}\int|f(\mathbf{q},t)|^2\exp(i\mathbf{qr})d\mathbf{q}. \tag{23}$$

Therefore the PSD is related to the Fourier transform of the surface topography.

$$S(\mathbf{q},t)=L^2|g_{nm}|^2. \tag{24}$$

For isotropic surfaces defined by the discrete set of height data the correlation function is given by

$$C_m=2\pi\sum_{k=0}^{N/2}k|g_k|^2J_0\left(2\pi\frac{km}{N}\right), \tag{25}$$

where $|g_k|^2$ and C_m are the angular averages of $|g_{nm}|^2$ and C_{kl}, respectively.

The width w of the thin film interface defined as the root mean square (rms) roughness is of particular interest.

$$w^2=\frac{1}{N^2}\sum_{k=1}^{N-1}\sum_{n=1}^{N-1}h_{k,n}^2=2\pi\sum_{n=0}^{N/2}n|g_n|^2. \tag{26}$$

Below we will compare the measured value of the roughness with the roughness expressed in terms of the discrete PSD,

$$w^2=C_0=\frac{2\pi}{L^2}\sum_{k=0}^{N/2}kS_k,\quad S_k=S(q_k,t). \tag{27}$$

According to Eq. (23), the roughness given by Eq. (27) consists of two terms

$$w^2=\frac{2\pi}{L^2}\sum_{k=0}^{N/2}kS_0(q_k)e^{-2a(q_k)t}+\frac{2\pi}{L^2}\sum_{k=0}^{N/2}kS_\eta(q_k,t). \tag{28}$$

The first term shows that the effect of initial roughness decreases with time. In physics terms this means that the information about the initial roughness is lost as the incoming atoms cover up the substrate. The second term is the roughness of the film grown on the initially flat substrate. It is known that this term is an increasing function of time.[14,16,18] The fact that the total width is the sum of the decreasing and increasing terms implies the possibility of nonmonotonic evolution, as has been indicated in Ref. 18. For sufficiently large systems $L\gg L^*$ in the limit of large times $t\gg D_4/D_2^2$ the Edwards–Wilkinson term dominates in Eq. (1).[18] This means that in this roughening regime the contribution to the rms roughness due to deposition noise [the second term in Eq. (28)] increases logarithmically with time.[15] Because of logarithmic dependence the transition from decreasing to increasing behavior of roughness is difficult to detect experimentally.

As we will see below the time dependence of the rms slope $\langle(\nabla h)^2\rangle^{1/2}$ provides some useful information about the interface smoothening process. The rms value of the interface slope within the sampling area can be calculated directly or using the Fourier transform

$$\langle(\nabla h)^2\rangle=\frac{1}{N^2}\sum_{k=1}^{N-1}\sum_{n=1}^{N-1}\left[\left(\frac{h_{k,n}-h_{k-1,n}}{\Delta x}\right)^2+\left(\frac{h_{k,n}-h_{k,n-1}}{\Delta y}\right)^2\right]=\frac{(2\pi)^3}{L^2}\sum_{k=0}^{N/2}|g_k|^2k^3, \tag{29}$$

where $\Delta x=\Delta y$ is the spacing between the sampling points. Then the experimental value can be compared with the model results.

$$\langle(\nabla h)^2\rangle=\frac{(2\pi)^3}{L^4}\sum_{k=0}^{N/2}k^3S_k. \tag{30}$$

The rms interface slope is averaged over all interface points and shows how deep are valleys on average as compared to valley widths.

B. Deposition noise strength

An exact calculation of the deposition noise strength is hardly possible. However it can be estimated using simple arguments. The growth rate of the coating consists of two contributions: (i) deposition of C atoms and (ii) film sputtering by Ar ions $F=F_+-F_-=(\alpha_C j_C^+-j_C^-)\Omega$, where Ω is the mean volume per atom in the film and j_C^+ and j_C^- represent the flux of carbon atoms to the substrate and resputtered from the substrate, respectively.

The sticking coefficient $\alpha_C<1$ (the fraction of collisions that result in the capture of carbon atoms by the thin film).[19] The maximum deviation of the growth rate from the average value is about the growth rate itself $|\max(\eta)|\sim F_+$; therefore, in the relation $\langle\eta(\mathbf{r},t)\eta(\mathbf{r}',t')\rangle=D\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$, the term in the left hand side is about F_+^2. In the right hand side (RHS), the combination $\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')$ is estimated as $\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')\sim\Omega^{-2/3}\tau_0^{-1}$, where $\Omega^{2/3}$ is the minimal "atomic area" and $\tau_0\sim\Omega^{1/3}F_+^{-1}$ is the average time interval between arrivals of atoms to that minimal area occupied by a single atom. Hence the strength of the deposition noise is proportional to the thin film growth rate,

$$D=F_+\Omega. \tag{31}$$

More complicated derivations can be found in Refs. 14 and 20. Resputtering of a growing thin film due to concurrent

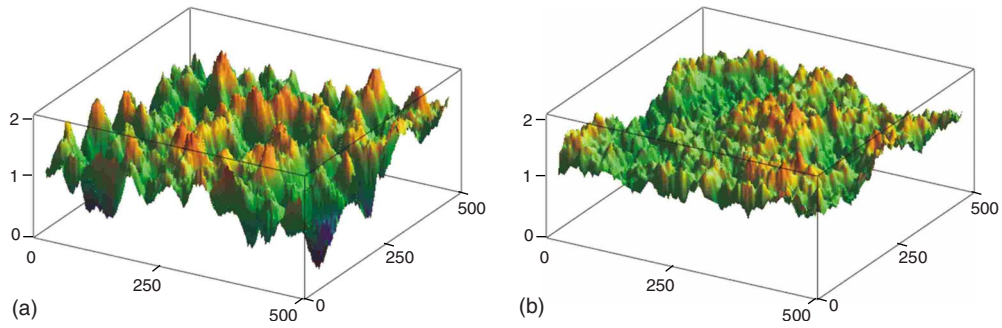

FIG. 1. (Color online) Typical examples of surface morphology: (a) the initial surface of the CrTi interlayer and (b) the surface of the TiC/*a*-C coatings after 4 h deposition hours in *p*-dc mode at 350 kHz. Height and lateral dimensions are given in nanometers.

ion impingement is statistically independent of deposition. Resputtering is equivalent to the arrival of atoms of "antimatter;" therefore resputtering contributes additively to the growth noise,

$$D = F_+\Omega + F_-\Omega = (\alpha_C j_C^+ + j_C^-)\Omega^2. \quad (32)$$

This expression shows that the optimum parameters of the *p*-dc power supply should be selected in order to avoid resputtering of the thin film and, at the same time, to ensure sufficient mobility of surface atoms due to the impingement of Ar^+ ions.

C. Estimation of diffusion coefficient D_2

As with any deposition method, the growth of a film is driven by a random arrival of atom/ions onto the substrate. If the adatoms have no or only limited possibility to migrate along the surface, the deposition noise inevitably causes a rapid increase in the interface roughness as a function of film thickness.[14,15] In principle, atomic mobility along the surface can be enhanced by elevation of the deposition temperature. However, it was found experimentally that dense DLC-based coatings with a high fraction of sp^3 atoms can only be formed if the deposition is carried out below a critical temperature of around 200 °C.[1,7] It is also known that the surface roughness increases if the deposition temperature is beyond the critical temperature (see, for example, the results of mass selected ion beam deposition[7]). Keeping the temperature below the critical temperature prevents the thermally activated processes in the bulk of DLC film and thus the formation of equilibrium carbon material (graphitization). In this case, a mechanism is necessary to increase the atomic mobility along the surface and in thin subsurface layer. This can be done by selecting the energy of depositing ions in an appropriate range and/or by concurrent impingement with Ar^+ ions, which may result in atomic rearrangements inside the subsurface layer of thickness comparable to the ion penetration depth. Low deposition temperature and strong atomic mixing induced by ion impingement, as the situation simulated in this paper, restrain possible epitaxial effects. As it was found in our experiments, orientation preference is not relevant to the growth behavior of TiC/*a*-C nanocomposite films where the amorphous carbon matrix dominates. High-resolution transmission electron microscopy observations revealed that TiC nanocrystallites isolated by amorphous carbon matrix do not exhibit any preferential orientation but rather orient randomly.[21] Nevertheless, preferential growth and/or suppression of growth of certain atomic planes may influence the roughness evolution of multicrystalline films, which is out of the scope of this paper.

The smoothening of initially rough interface during deposition of ta-C has been explained in Ref. 22 by local melting in the region of impact-induced thermal spikes that result in a reduction in local surface curvature. However, in Ref. 23 on the basis of MD simulations[19] it was noticed that the size and duration of a thermal spike in amorphous carbon seem too small for the establishment of a liquidlike behavior. Instead, the smoothness of DLC thin films deposited with carbon ion beam has been explained by ballistic effects resulting in downhill diffusion along the inclined surface.[23] This mechanism is likely relevant to our case of *p*-dc magnetron sputtering deposition. From MD simulations the authors of Ref. 23 concluded that Edwards–Wilkinson diffusion flux along the interface is given by

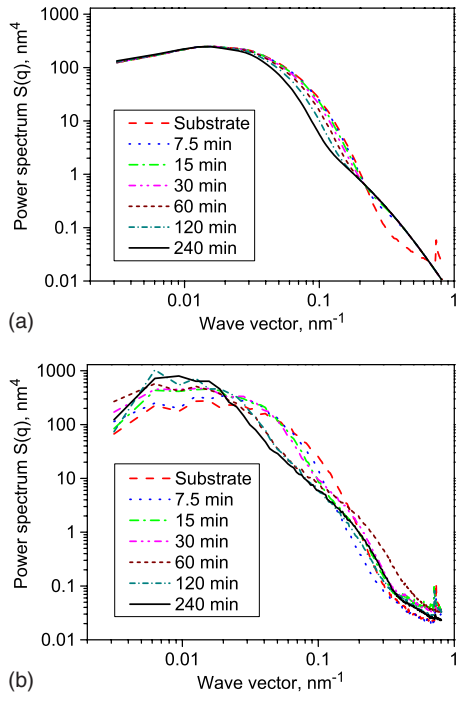

FIG. 2. (Color online) Comparison of (a) the model PSD with (b) the PSD obtained from the experimental data.

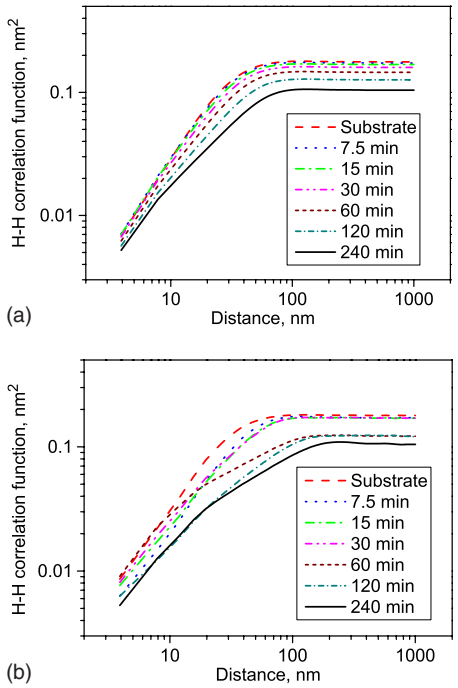

FIG. 3. (Color online) Height-difference correlation function $G(\mathbf{r})=\langle[h(\mathbf{r}+\mathbf{r}')-h(\mathbf{r}')]^2\rangle=2[C(0)-C(\mathbf{r})]$. Comparison of the model predictions (a) with the experimental data (b).

$$\mathbf{j}_2 = -J_C \Omega \nu(E) \nabla h = -D_2 \nabla h, \tag{33}$$

where J_C is the carbon-ion flux and $\nu(E)$ is the smoothening strength that is proportional to the sum of lateral atomic displacements in the vicinity of an energetic particle impinged on the thin film. According to Ref. 23, $\nu(E)$ increases linearly with the energy of incoming carbon ions up to the value of about 2 nm at $E^* \approx 120$ eV, then it saturates because for higher energies ions penetrate into the subsurface layer and release a part of the impact energy in the bulk.

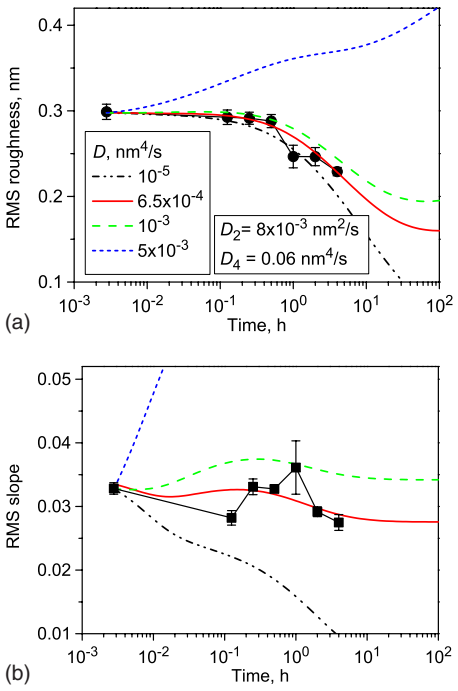

FIG. 4. (Color online) Time dependence of (a) the rms roughness and (b) the rms slope at several values of the noise strength D. The model results are compared with the experimental data shown by full circles.

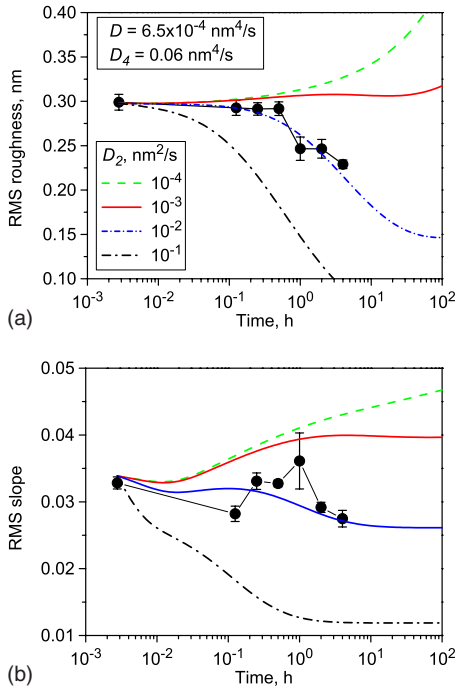

FIG. 5. (Color online) Time dependence of (a) the rms roughness and (b) the rms slope at several values of the parameter D_2.

In the case of p-dc magnetron sputtering deposition, the Edwards–Wilkinson diffusion coefficient can be written in terms of Ar^+ flux to the growing thin film,

$$D_2 = \Omega \int \nu(E) f_{\mathrm{Ar}}^S(E) dE = b\Omega \int_0^{E^*} E f_{\mathrm{Ar}}^S(E) dE + bE^*\Omega \int_{E^*}^{\infty} f_{\mathrm{Ar}}^S(E) dE. \tag{34}$$

Here the energy distribution of Ar^+ ion flux to the substrate is

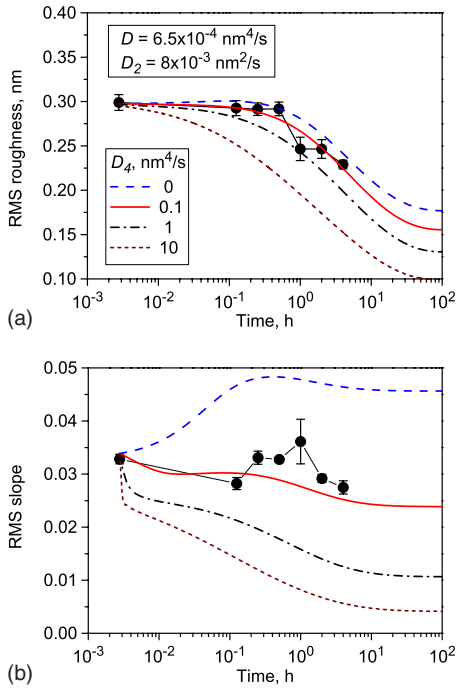

FIG. 6. (Color online) Time dependence of (a) the rms roughness and (b) the rms slope at several values of the parameter D_4.

denoted as $f_{\mathrm{Ar}}^{S}(E)$ and $\nu(E)$ can be approximated by the dependence

$$\nu(E) = \begin{cases} bE, & E < E^*, \\ bE^*, & E > E^*, \end{cases} \tag{35}$$

where b is a constant independent of the energy of impinging ions.

The first term in the RHS of Eq. (34) is proportional to the total energy delivered by the low energy Ar^+ ions to the subsurface layer of the thin film. The second term shows that only a fraction of energy of the energetic ions is available to induce diffusion along the surface. The rest of the energy is released in the bulk of the thin film.

D. Estimation of diffusion coefficient D_4

Following Mullins,[17] the kinetic coefficient D_4 is usually attributed to the interface diffusion of adatoms driven by the gradient of the chemical potential along the interface, which is associated with the gradient of the local curvature. Under near-equilibrium conditions the Mullins expression for the coefficient D_4 is given by

$$D_4 = \frac{D_s \gamma \Omega^2 n}{k_B T}, \tag{36}$$

where D_s is the coefficient of interface diffusion, γ is the interface energy, and n is the number of interface atoms per unit area. In the case of nonequilibrium process of crystal growth (molecular beam epitaxy) the number of interface atoms is determined by the balance between the arrival rate of incoming atoms and the rate of their absorption by steps; the step density is determined self-consistently by kinetics of nucleation and attachments of adatoms to step edges. For amorphous thin films it is not clear how to define the number of interface atoms or the number of interface defects that absorbs arriving atoms.

III. COMPARISON WITH THE EXPERIMENTAL DATA

TiC/*a*-C nanocomposite thin films were prepared in the closed-field unbalanced magnetron sputtering system Teer UDP 400/4, which operated in pulsed-dc mode at 350 kHz.[12] Substrates were not intentionally heated during deposition. The substrate temperature was measured to be 190 °C, just below the critical temperature of sharp fall of sp^3 fraction.

The total fraction of carbon atoms in the film of optimum composition was about 68 at. %. Prior to depositing TiC/*a*-C thin films the CrTi film was deposited onto the ⌀100 mm Si wafer. A Veeci NanoScope IIIa atomic force microscope (AFM) was used to image the surface morphology and to measure the surface roughness of 2×2 μm^2 samples.

In order to investigate the smoothening kinetics, a set of thin film samples was deposited for different times. The maximum deposition time was 4 h and the deposition rate was about 0.1 nm/s. Figure 1 demonstrates an obvious reduction in interface roughness of TiC/*a*-C thin films with deposition time.

To compare the simulated result with the experimental data, the deposition noise [Eq. (32)] was fixed at the value $D = 6.5 \times 10^{-4}$ nm^4/s that corresponds to the mean atomic volume $\Omega = 6.5 \times 10^{-3}$ nm^3. The parameters D_2 and D_4 were adjusted to obtain a good fit to experimental data. The fitting procedure was as follows. (i) The angular average of the initial PSD $S_0(q_k)$ was restored using the AFM measurements in 512×512 points of the initial interface of the CrTi interlayer [see Eq. (24)]: this PSD was averaged over eight samples. (ii) Then D_2 and D_4 were evaluated by comparing the time dependencies of PSD, rms roughness, and rms slope [Eqs. (19), (27), and (30)] with the corresponding experimental dependencies. A good agreement with experimental data was obtained at $D = 6.5 \times 10^{-4}$ nm^4/s, $D_2 = 8 \times 10^{-3}$ nm^2/s, and $D_4 = 0.06$ nm^4/s (Figs. 2 and 3).

At given "smoothening efficiency", D_2 and D_4, the roughness and the mean slope of the thin film strongly depend on the deposition noise strength D (Fig. 4). It has been shown in Sec. II that the deposition noise is proportional to the thin film growth rate; therefore, it cannot be removed completely. Roughness can be controlled effectively by increasing the mobility of adatoms with concurrent ion impingement. Figure 5 shows the influence of the diffusivity parameter D_2 on the time dependence of rms roughness and rms slope.

Roughness evolution is less sensitive to the variation in the parameter D_4 (Fig. 6) as compared to that of the parameter D_2. This means that the Edwards–Wilkinson relaxation term dominates in the process. However the fourth-order term is necessary in order to reproduce the shape of the PSD and the rms slope observed experimentally.

IV. CONCLUSIONS

In this work, we have presented an analysis of smoothening mechanism of the thin film grown with *p*-dc magnetron sputtering. Roughness evolution has been described by the linear stochastic equation, which contains the second- and fourth-order gradient terms. The origin of the second-order Edwards–Wilkinson terms can be explained by ballistic effects resulting in downhill diffusion along the inclined surface.[23] Ballistic effects are sensitive to the flux and energy of Ar^+ ions that impinge the growing thin film. Therefore the thin film roughness can be controlled by adjusting the waveform, frequency, and width of dc pulses. The model results well agree with the experimental data.

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